

AIR QUALITY TRENDS MODELLING IN EUROPE OVER THE PAST DECADE

Augustin Colette¹, Bertrand Bessagnet¹, Ariela D'Angiola², Michael Gauss³, Claire Granier², Øivind Hodnebrog⁴, Hermann Jakobs⁵, Alberto Maurizi⁶, Frédérik Meleux¹, Michael Memmesheimer⁵, Agnes Nyiri⁷, Laurence Rouil¹, Felicità Russo⁶, Sverre Solberg⁸, Frode Stordal⁴, Francesco Tampieri⁶

¹ INERIS, BP 2, 60550 Verneuil-en-Halatte, France

² CNRS/IPSL, France

³ MET.NO, Norway

⁴ UiO, Norway

⁵ FRIIUK, Germany

⁶ CNR/ISAC, Italy

⁷ EMEP

⁸ NILU, Norway

augustin.colette@ineris.fr

Introduction

Atmospheric Chemistry Transport Models (CTM) are essential tools in the design and evaluation of air quality (AQ) management policies. Making use of sensitivities studies, where anthropogenic emissions fluxes are artificially perturbed in model simulations, one can identify the best practices in terms of air pollution control. However if such models are now well established and being implemented by a wide community of research, governmental or private institutions, their validation against measurements is usually limited to temporal snapshots: isolated pollution episodes or single-year validation periods.

We propose a dynamic evaluation of state-of-the-art models by discussing their ability to capture air pollution changes over the past decade. Six modeling teams were involved in the experiment: two of them operating global models while the others used regional (limited area) models. The focus of the study was limited to Europe over the 1998-2007 decade. In the present paper, after an overview of the observed AQ trends over Europe, we introduce the modeling experiment and highlight the main findings in a last section. More information about this activity can be found in Colette et al. (2011).

Observed AQ trends over Europe

In order to address trends in air quality posing a threat on human health exposure, the observational atmospheric chemistry data used in the present study were obtained from the AIRBASE repository of European air quality monitoring networks that include urban, suburban and rural stations. This way we could study the capability of models to capture trends in polluted places as opposed to background locations, documented for instance by the EMEP network of remote stations. The drawback of using the AIRBASE dataset is that the stations are designed for exposure assessment rather than trend studies, so that an important data cleansing procedure had to be completed. Using only the stations complying to the data coverage and completeness criteria, we found the trends represented on Figure 1 for NO₂, O₃ and PM₁₀. These trends are computed using time series of monthly averages of daily mean values at each individual location. Each record is de-seasonalised by removing the average seasonal cycle from the raw monthly record and the slope is then computed using a standard linear least square method. The significance of the trend is assessed with a Mann-Kendall test at the 95% confidence level to account for auto-correlation and seasonality.

The decrease of NO₂ concentration is quite robust throughout Europe, except in South-Eastern France and Northern Italy plus a couple of isolated stations. It appears on these maps that the average trend is more pronounced at urban stations: the median trend for all UB (urban background), SB (suburban background) and RB (rural background) stations are -0.37, -0.27 and -0.14 µg/m³/year, respectively. We find an absolute majority of European stations with a significant negative trend: 62%, 52% and 53% (UB, SB and RB).

These decreasing trends for nitrogen dioxide are reflected in the evolution of O₃ where a slight increase is observed especially at urban sites because anthropogenic emissions are high enough so that a decrease of NO_x has primarily an impact on the reduction of night-time titration. Hence we find that the average daily mean O₃ trend at UB, SB and RB sites is 0.37, 0.27 and 0.05 µg/m³/year, respectively.

The number of PM10 monitoring stations that pass the data completeness criteria is by far lower than for O₃ or NO₂. In Germany, UK, and Benelux, PM10 concentrations are systematically decreasing, thanks to the air quality regulation enforced during the past decade. However, the trend of total PM10 levels-off in Northern Germany and the UK. In parts of Spain and the Czech Republic, a positive trend is found.

Multi-model experiment

Setup

In order to produce an ensemble of models that best represents our ability to capture air quality trends, it was decided to keep the

modelling setup as flexible as possible, the only restriction being to use the same emission inventory for anthropogenic emissions. As such, the present experiment is not a model inter-comparison initiative, but rather an attempt to assess the uncertainties in air pollution trend modelling.

The anthropogenic trend was that of the EMEP emission inventory (Vestreng et al., 2005) which is based on official emission data reported by individual countries under the LRTAP convention. Beyond the European domain (for global CTMs), these emissions are merged into the MACCity inventory (Granier et al., 2011).

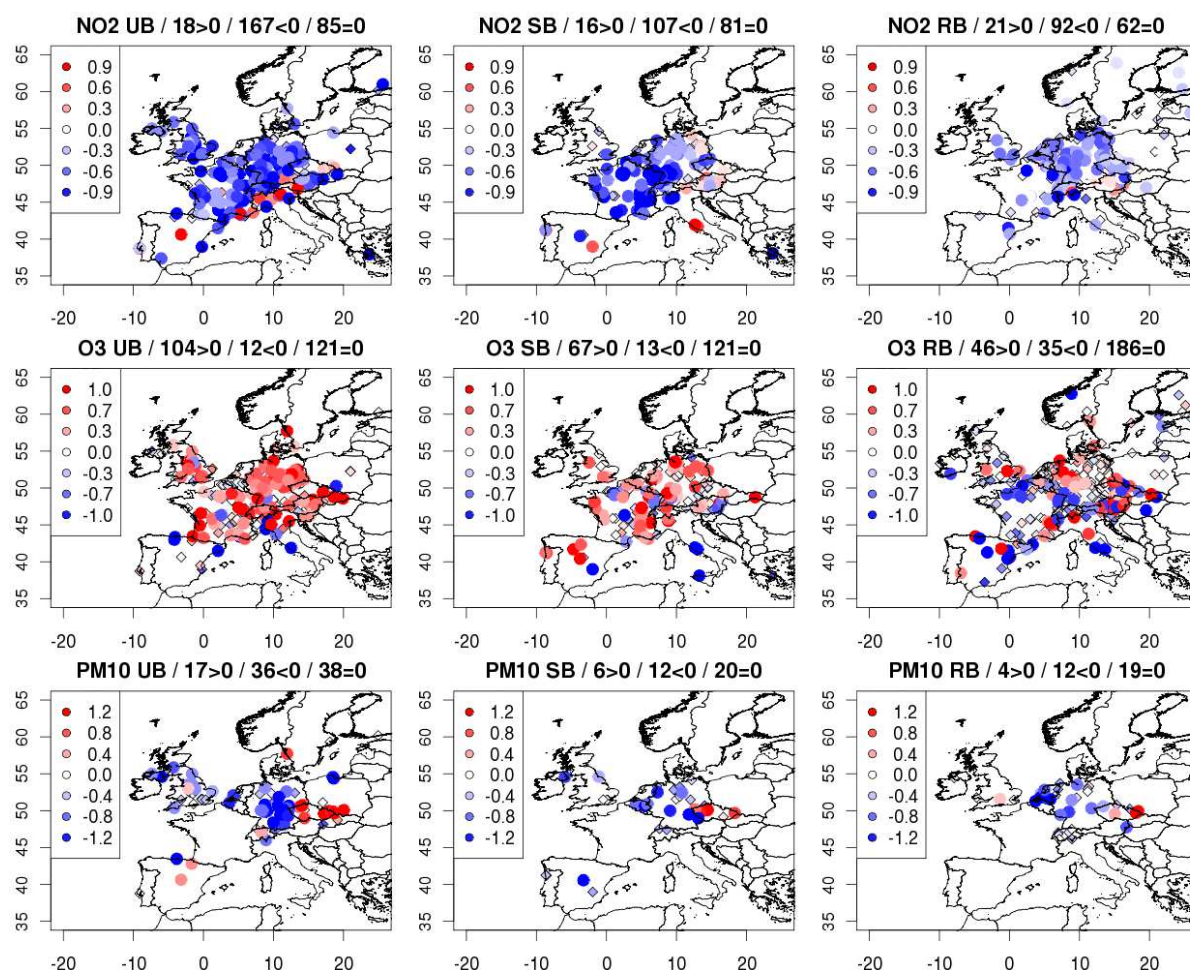


Figure 1: Trends of daily means of NO₂, O₃, and PM₁₀ (µg/m³/yr) observed at urban background (UB), suburban background (SB) and rural background (RB) AIRBASE stations. Stations where a statistically significant trend is observed are shown with a large dot a small diamond is used otherwise. The title of each panel also provides the number of stations with a positive, negative or null (not significant) slope. From Colette et al. 2011.

Six CTMs simulated the whole 10 years over Europe at a resolution of about 50km for the regional models, and 2 to 3 degrees for the global models. Specifically the regional CTMs involved were: Bolchem (Mircea et al., 2008), Chimere (Bessagnet et al., 2008), Emep (Simpson et al., 2011), Eurad (Memmesheimer et al., 2007) and the global models were: OsloCTM2 (Isaksen et al., 2005) and Mozart (Emmons et al., 2010).

Modeled trends

The NO₂ trends (see Figure 3 of Gauss et al., this issue) are very tightly constrained by the evolution prescribed in the emission inventory. When considering only stations where a significant NO₂ trend is measured the sign of the trend is well captured at 73% ($\sigma = 6\%$) of the sites on average across all models. The main discrepancies are clearly correlated with country boundaries pointing towards uncertainties in the reported emission inventories.

For Ozone, the observed trends are much milder than for NO₂. This feature, in addition to the less linear formation process makes it more challenging for the models to capture. We display on Figure 2 the modeled trend for O₃. Note that here the trend is reported at every grid point with no regards to its significance. The most consistent feature over land surfaces is a widespread decrease of O₃, except in the hotspot of NO_x emissions constituted by the UK-Benelux-Germany area because of the NO_x-saturated character of this region, emphasizing the titration effect of NO_x emission reduction over the past 10 years. This feature is captured by regional and global models whereas the later were not thought to capture such specificities given their horizontal resolution. Only Eurad produces a slightly different picture.

The comparison with the trends reported at Airbase stations shows that the picture reflected by the models matches more closely

the trends at rural stations with positive slopes over the greater Benelux area and a decreasing ozone elsewhere (over continental surfaces). So that the slight increase in ozone observed throughout Europe at urban and suburban sites as a result of the lower titration induced by the reduction of NO_x emissions is not captured by the models. This feature is not surprising given the resolution of the models involved in this experiment. It is nevertheless an important limitation of the study since we cannot conclude on the skillfulness of the models to capture trends in urban areas outside of the Benelux area.

Conclusion

We conducted a first multi-model assessment of air quality trends at the regional scale. Six chemistry transport models were implemented over the 1998-2007 decade over Europe and compared to AQ monitoring stations. The capability of the models to capture trends of primary and secondary constituents was assessed showing that the slight increase of ozone observed in urban areas over the past decade could only be reproduced robustly over the Benelux area.

This initiative led also to an assessment of trends in particulate matter. And the experiment was repeated with constant emissions for 4 out of the 6 models in order to investigate the respective role of emissions and meteorology on AQ trends.

More details can be found in a recently published full length paper (Colette et al. 2011).

Acknowledgment

The research leading to these results has received funding from the European Union's Seventh Framework Programme (FP7/2007-2013) for the CityZEN Project, under grant agreement no. 212095.

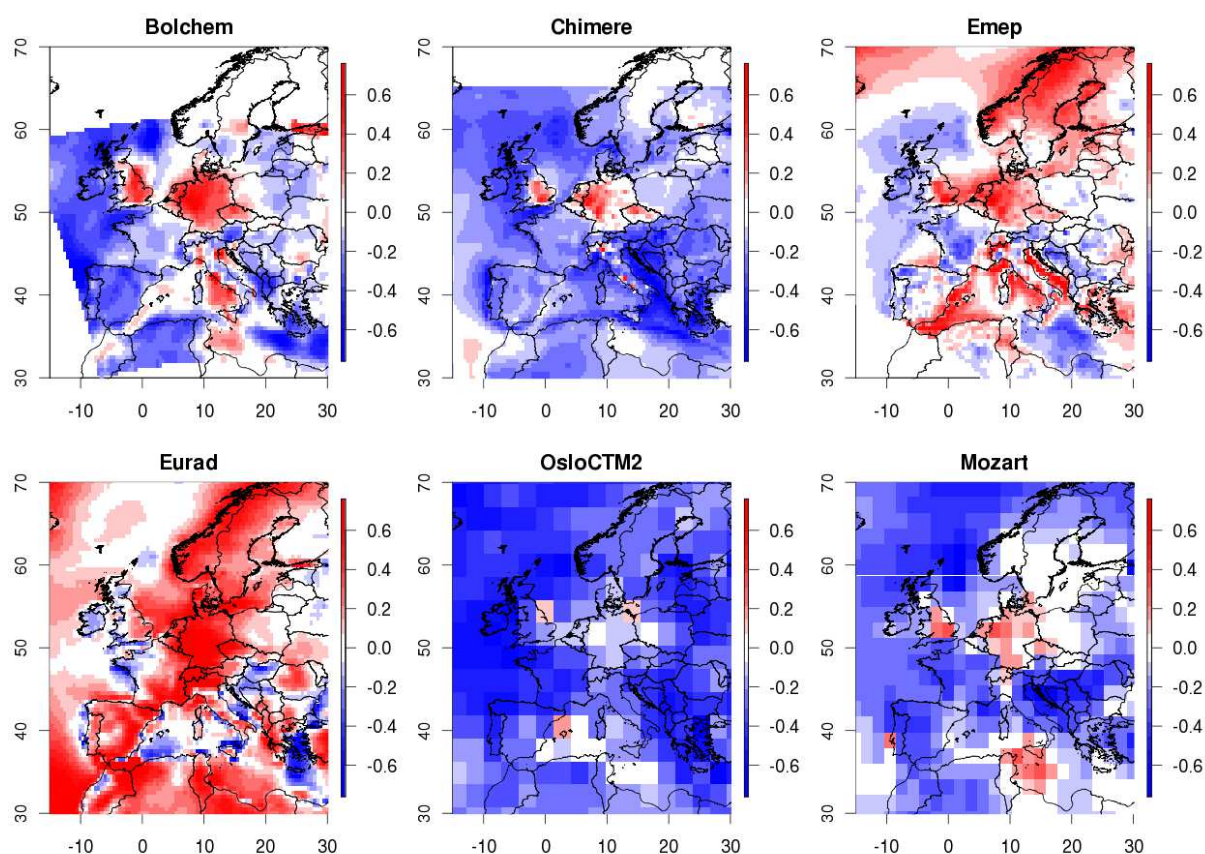


Figure 2: Modelled O₃ trend (µg/m³/yr) for each CTM and at each grid point computed on the basis of monthly means of daily means over the 1998-2007 period with a linear least square fit of de-seasonalised values. From Colette et al. (2011).

References

Bessagnet, B., Menut, L., Curci, G., Hodzic, A., Guillaume, B., Liousse, C., Moukhtar, S., Pun, B., Seigneur, C., and Schulz, M.: Regional modeling of carbonaceous aerosols over Europe—focus on secondary organic aerosols, *Journal of Atmospheric Chemistry*, 61, 175-202, 2008.

Colette, A., et al., 2011: Air quality trends in Europe over the past decade: a first multi-model assessment, *Atmos. Chem. Phys. Discuss.*, 11, 19029-19087.

Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J. F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L., and Kloster, S.: Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), *Geosci. Model Dev.*, 3, 43-67, 2010.

Gauss, M. Environmental Impact of Megacities - Results from CityZen. Proceeding of the Accent-plus symposium, Urbino Italy 2011.

Granier, C., Bessagnet, B., Bond, T., D'Angiola, A., Denier van der Gon, H., Frost, G., Heil, A., Kaiser, J., Kinne, S., Klimont, Z., Kloster, S., Lamarque, J.-F. o., Liousse, C., Masui, T., Meleux, F., Mieville, A., Ohara, T., Raut, J.-C., Riahi, K., Schultz, M., Smith, S., Thompson, A., van Aardenne, J., van der Werf, G., and van Vuuren, D.: Evolution of anthropogenic and biomass burning emissions of air pollutants at global and regional scales during the 1980-2010 period, *Climatic Change*, 109, 163-190, 2011.

Isaksen, I. S. A., Zerefos, C., Kourtidis, K., Meleti, C., Dalsøren, S. B., Sundet, J. K., Grini, A., Zanis, P., and Balis, D.: Tropospheric ozone changes at unpolluted and semipolluted regions induced by stratospheric ozone changes, *J. Geophys. Res.*, 110, D02302, 10.1029/2004JD004618, 2005.

Memmesheimer, M., Wurzler, S., Friese, E., Jakobs, H. J., Feldmann, H., Ebel, A., Kessler, C., Geiger, J., Hartmann, U., Brandt, A., Pfeffer, U., Dorn, H. P., and Carlos Borrego and Eberhard, R.: Chapter 2.8 Long-term simulations of photo-oxidants and particulate matter over Europe with emphasis on North Rhine-Westphalia, in: *Developments in Environmental Sciences*, Elsevier, 158-167, 2007.

Mircea, M., D'Isidoro, M., Maurizi, A., Vitali, L., Monforti, F., Zanini, G., and Tampieri, F.: A comprehensive performance evaluation of the air quality model BOLCHEM to reproduce the ozone concentrations over Italy, *Atmospheric Environment*, 42, 1169-1185, 2008.

Simpson, D., Benedictow, A., Berge, H., Fagerli, H., Gauss, M., Jonson, J. E., Nyiri, A., Semeena, V., Steensen, B. M., Tsyro, S., Valdebenito, A., and Wind, P.: The EMEP MSC-W Chemical Transport Model I: Model Description, in preparation, 2011.

Vestreng, V., Breivik, K., Adams, M., Wagener, A., Goodwin, J., Rozovskaya, O., and Pacyna, J. M.: Inventory Review 2005, Emission Data reported to LRTAP Convention and NEC Directive, Initial review of HMs and POPs, MSC-W, 2005.